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Arc Plasma Synthesis of Nanostructured Fe Powder

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ABSTRACT

To investigate the effect of the parameters of the arc plasma process on the characteristics of the iron nano powder, the hydrogen volume fraction in the powder synthesis atmosphere was changed from 10% to 50%. The particle size, phase structure and magnetism of the synthesized iron powder were studied using the FE-TEM, XRD and a vibration magnetometer at room temperature. The particle size increased simultaneously with the increase in the hydrogen volume fraction, and the particle size ranged from about 20nm to 100nm with the change of the hydrogen volume fraction increasing from 10% to 50%. The synthesized iron powder particle had a shell-core structure composed of the crystalline α -Fe in the core and the crystalline Fe_3O_4 in the shell. The iron nano powder synthesized under the high hydrogen volume fraction condition had a higher saturation magnetism and lower coercive force than the values of the one synthesized under the low hydrogen volume fraction condition.

INTRODUCTION

Metal nano particles with a diameter between 10nm and 100nm have a wide range of potential application, including magnetic recording media, catalyst, electric conductive or resistive paste, ferrofluids, and others [1]. The iron is one of the main elements for the application of metal nano particles in the various industrial fields. Nano size iron particles are of particular interest in the fundamental studies of magnetism. The magnetic properties of nano sized iron particles, prepared by varied methods at varied conditions are very different [2,3]. The physical and chemical properties of metal nano particles strongly depend on the particle and surface properties, including size, morphology, surface area, surface oxide, and others. The practical application of metal nano particles should be to verify the relationship between the preparing process parameters and those properties.

Nano particles can be produced by several different methods, such as colloidal precipitation, mechanical attrition and the gas condensation process [4]. The latter is the most used method at present, for the advantages of being relatively simple to be extended to large-scale and high-rate production. The gas condensation processes typically involve nucleation and

growth of nano sized particles from a supersaturated vapor produced either by evaporation of bulk materials or by chemical reaction of gas-phase precursors. The arc plasma process can be preparing metal, ceramic and their composite nano particles by vaporization-condensation of metals or alloys in an active atmosphere using a DC arc-plasma. The nano particles prepared by this process are well dispersed and free of anion contamination compared to those prepared by the wet process.

In the present study, nano iron particles were prepared by the arc plasma process with changing process parameters, and then the morphology, phase structure and magnetism of synthesized iron particles at different process parameters were studied by using the FE-TEM, XRD and a vibration magnetometer at room temperature.

EXPERIMENTAL DETAILS

The preparing apparatus of nano particles by the arc plasma method mainly consisted of a vacuum chamber, tungsten cathode and a copper anode, a gas flow system and a DC power supply. The pure iron (above 99.99% purity) bulk was used as a raw material. The vacuum chamber was pumped to 10^{-5} torr and then was backfilled with a hydrogen/argon mixture gas to 300 torr. Then the arc plasma was initiated between the tungsten cathode and the iron bulk on the copper anode under the arc current 60A, arc voltage 21~25V, and a reaction time of 1 hour. The mixing ratio of the hydrogen/argon was changed from 1:9 to 5:5. The atom smoke of the iron was subsequently generated around the arc spot area of the molten iron. The nano iron particles were formed by a collision between the quenched iron atoms due to a collision with atmosphere gas atoms. Then the formed nano iron particles were separated with working gas. Before the particles were taken out of the chamber, they were passivation treated under an argon/oxygen mixture atmosphere at room temperature for 12 hours to prevent a rapid spontaneous combustion. The variation of microstructure, phase structure and magnetic properties of synthesized iron particles with the change of the mixing ratio of the hydrogen/argon were studied using the FE-TEM, XRD and a vibration magnetometer at room temperature.

RESULTS AND DISCUSSION

Figure 1 shows X-ray diffraction patterns of synthesized powder with the volume fraction of hydrogen gas in the powder synthesis atmosphere. In the case of a 10vol.% of hydrogen, synthesized powder was composed of α -Fe and Fe_3O_4 phases. The relative intensity of the diffraction peak of Fe_3O_4 decreased with an increase in the volume fraction of hydrogen, and the diffraction peak of Fe_3O_4 disappeared at above about a 30vol.% of hydrogen. The Fe_3O_4 phase

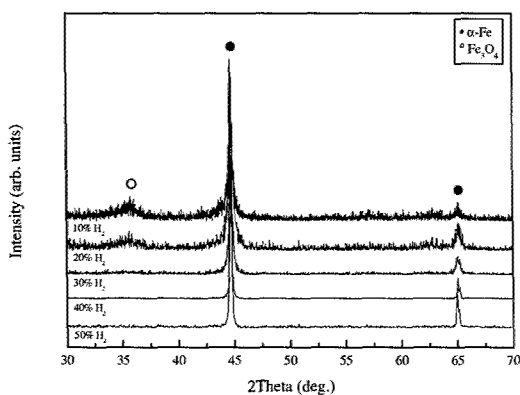


Figure 1. The variation of X-ray diffraction patterns of synthesized powder with the volume fraction of hydrogen.

was formed due to the surface oxidation reaction of the iron particles by the oxygen atoms during the passivation treatment. The surface oxidation reaction of the particles depends on the particle size and the relative amount of the particle to the oxygen concentration in the passivation atmosphere. Figure 2 shows the variation of the powder generation rate with the volume fraction of the hydrogen in the powder synthesis atmosphere. The powder generation rate slowly increased with an increasing of hydrogen volume fraction until at about a 20vol.%, and then dramatically increased at about a 30vol.% of hydrogen, and then had saturated values about 3.5g/h at above a 40vol.% of hydrogen. By the Uda [5], the hydrogen molecular promotes the evaporation of the metal vapor from the molten metal in the arc plasma process by the dissolving and evaporation action. This promoting action of the hydrogen molecular increased with increasing the volume fraction of the hydrogen in the powder synthesis atmosphere by Sievert's

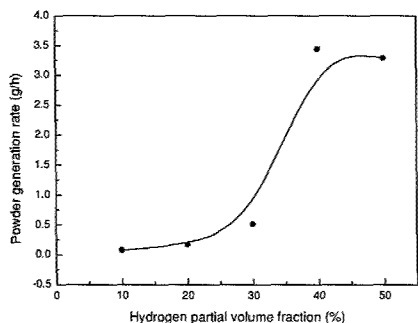


Figure 2. The variation of the powder generation rate with the volume fraction of hydrogen.

law. Also, the powder generation rate increases with increasing the melt temperature at a constant chamber pressure. The thermal energy of the arc plasma depends on the gas atmosphere and shape of one's column. The shape of the arc plasma column that was changed from a spread shape has low thermal energy to a concentration shape and has a high thermal energy with increasing of the volume fraction of the hydrogen. It knows that the synthesis condition was changed from a small amount of powder generation condition to a large amount of powder generation condition with increasing of the volume fraction of the hydrogen in the powder synthesis atmosphere. The oxygen concentration in the chamber was constantly maintained during the passivation treatment. The high powder generation rate on the powder synthesis step has an effect of increasing the relative amount of powder to oxygen concentration on the passivation treatment step. Figure 3 shows the TEM micrographs of the synthesized powder with the hydrogen volume fraction of (a) 20% and (b) 40%. As shown in figure 3, the particle size has about 20nm at a 20vol.% of hydrogen, and has about 80nm at a 40vol.% of hydrogen. It knows that the particle size of the synthesized iron powder increased with increasing of the volume fraction of the hydrogen. As shown in figure 2, the powder generation rate increased with an increasing of the volume fraction of the hydrogen. This means that the concentration of the iron vapor in the chamber was increased with an increasing of the volume fraction of the hydrogen. The higher iron vapor concentration gives a higher collision probability between the cooled iron atoms, and then formed larger iron particles. The larger particle has less sensitivity on oxidation than the smaller particle due to the relatively small surface area per unit volume. It was considered that the phase change of the synthesized iron powder with the hydrogen volume fraction as shown in figure 1, was due to the change of the relative amount of the powder generation and particle size with the hydrogen volume fraction.

Figure 4 shows the FE-TEM micrograph of the synthesized iron nano particle at 20vol.% of the hydrogen. The synthesized iron nano particle had a shell-core structure composed of α -Fe

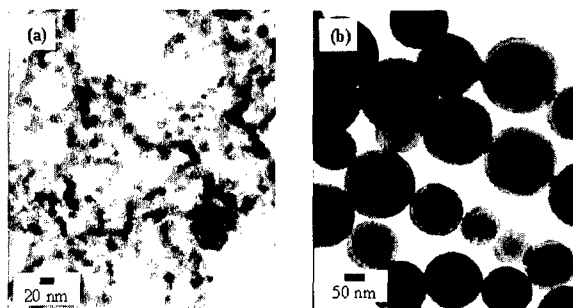


Figure 3. TEM micrographs of the synthesized powder with the hydrogen volume fraction of (a) 20% and (b) 40%.

in the core and Fe_3O_4 in the shell. The oxide layer has a crystalline structure, and the thickness of one layer was about 2nm at a 20vol.% of the hydrogen. The oxide thickness decreased from about 2.5nm to 1nm with increasing of the volume fraction of the hydrogen from 10% to 50%.

Figure 5 shows the magnetization hysteresis loops of the synthesized iron nano powder with the hydrogen volume fraction of (a) 20% and (b) 40%. In the case of a 20vol.% of the hydrogen, the saturation magnetization and coercive force have about 120emu/g and 750Oe, respectively. In the case of a 40vol.% of the hydrogen, the saturation magnetization and coercive force have about 180emu/g and 150Oe, respectively. The iron nano powder synthesized under the high hydrogen volume fraction condition had a higher saturation magnetism and lower coercive force than the values of the one synthesized under the low hydrogen volume fraction condition. It thinks that these changes of the saturation magnetization and coercive force of the synthesized iron nano powder comes from the change of the relative amount of composing phases ($\alpha\text{-Fe}$ and Fe_3O_4) and particle size with the volume fraction of the hydrogen [6,7]

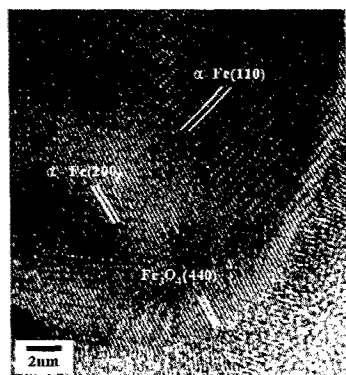


Figure 4. FE-TEM micrograph of the synthesized iron nano particle.

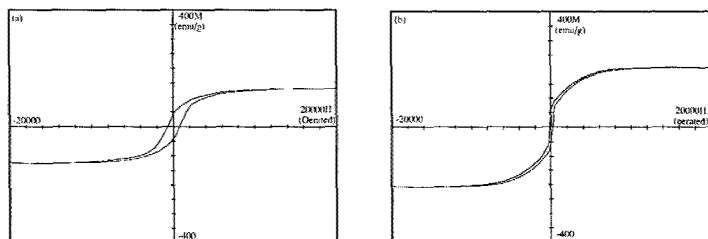


Figure 5. The magnetization hysteresis loops of the synthesized iron nano powder with the hydrogen volume fraction of (a) 20% and (b) 40%.

CONCLUSIONS

The iron nano powder could be synthesized by the arc plasma process and this powder has a stable surface characteristic on the rapid spontaneous combustion. The powder generation rate and particle size were increased with increasing of the hydrogen volume fraction in the powder synthesis atmosphere. The particle size ranged from about 20nm to 100nm with the change of the hydrogen volume fraction increasing from 10% to 50%. The synthesized iron powder particle had a shell-core structure composed of the crystalline α -Fe in the core and the crystalline Fe_3O_4 in the shell. The thickness of the oxide layer decreased simultaneously with the increase in the hydrogen volume fraction in the powder synthesis atmosphere. This thickness ranged from about 2.5nm to 1nm with the change of the hydrogen volume fraction increasing from 10% to 50%. The iron nano powder synthesized under the high hydrogen volume fraction condition had a higher saturation magnetism and lower coercive force than the values of the one synthesized under the low hydrogen volume fraction condition.

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REFERENCES

1. R. W. Siegel, *Mater. Sci. Eng.* **A168**, 189(1993).
2. Y. Fukano, *Jpn. J. Appl. Phys.* **13**, 1001(1974)
3. M. Kusunoki and T. Ichihashi, *Jpn. J. Appl. Phys.* **25**, 219(1986).
4. K. E. Gonsalves, S. P. Rangarajan and J. Wang in *Handbook of Nanostructured Materials and Nanotechnology* edited by H. S. Nalwa, (Academic Press 24-28 Oval Road, London, 2000) pp.1-52.
5. M. Uda, *Bull. Metal. Soc. Japn.* **22**, 412(1983).
6. T. A. Yamamoto, R. D. Shull and H. W. Hahn, *Nanostr. Mater.* **9**, 9530(1977).
7. A. E. Berkowitz, R. H. Kodama, S. A. Makhlof, Ft. Parker, F. E. Spada, E. J. McNiff and S. Foner, *J. Magnetism and Magnetic Mater.* **196-197**, 591(1999).